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PROCESS FOR THE CATALYTIC POLYMERIZATION OF OLEFINS, A REACTOR SYSTEM, ITS USE IN THE PROCESS, THE POLYOLEFINS OBTAINED AND THEIR USE.

The present invention relates to a process for the catalytic polymerisation of olefins, to a particular reaction system, to the use of this reactor system for the catalytic polymerization of olefins, to the polyolefins obtained by the said process and to the use of the polyolefins.

The catalytic polymerization of polyolefins uses a catalyst of the Ziegler-Natta type. New generations of catalyst for olefin polymerization including single site catalysts referred to as Ziegler-Natta catalysts have been developed in view of a higher activity and/or selectivity. During the catalytic polymerization the olefin composition is substantially constant around the particle. Reactor gasses exiting the reactor are compressed and recycled. Make-up monomers and optionally hydrogen are added as needed. Entrained particles can be separated by an interposed cyclone and recycled to the polymerization reactor.

Polyolefins produced using a recent generation of Ziegler-Natta catalysts have a relative narrow molecular weight distribution. The breadth of the molecular weight distribution influences the rheology behaviour and the final mechanical properties of the produced polyolefins.

In order to obtain a broader bimodal molecular weight distribution, use is made of different reactor conditions, such as different concentrations for monomer, comonomer and/or hydrogen. Another option is the use of specific and/or combined catalysts.

Traditionally, series of reactors are used for applying different reaction conditions during the catalytic polymerization in order to obtain a broad or bimodal molecular weight distribution.

The use of one or more fluidized bed reactors in combination with a liquid phase reactor is for example described in EP 0517 868. Herein the use of a multi-stage process for producing bimodal polyolefins and polyolefins with a broad molecular weight distribution is described. This multi-state process comprises a first reactor for carrying out a first polymerization reaction, a second reactor for carrying out a second polymerization and optionally a third reactor for carrying out a third polymerization. The first reactor is a loop reactor wherein olefins are polymerized in an inert hydrocarbon medium. The second and third reactor are fluidized bed reactors such as gas phase reactors. It is an object of the present invention to further improve a process like the mentioned process. In relation to a enhanced homogeneity, improvement of the molecular weight distribution resulting in an improvement of the strength of the product and a more even comonomer distribution. Possibly a second gas phase rector is no longer necessary.

A first aspect of the present invention relates to a process for the catalytic polymerization of olefins comprising the steps of;

i) a first polymerization in a first reactor, wherein olefins are polymerized with a particulate catalyst, hydrogen and optional a comonomer in a fluidum of an inert low boiling hydrocarbon medium into an reaction mixture comprising polymerized olefins; and

ii) a second polymerization in a second reactor, wherein the polymerized olefins are further polymerized in a fluidized bed and in a moving bed under such conditions that the residence time in the fluidized bed and the residence time in the moving bed are independently controlled.

With this process it is feasable to provide a variety of polyolefins, such as monomodal and bimodal products with an improved polydispersity and/or bimodality/multimodality. Further, it is possible to control the built up of the polyolefin particles, such that a desired number and thickness of the polymeric layers is obtained.

Preferably, the inert low boiling hydrocarbom medium comprises propane, butane, isobutane pentane, hexane, heptane, octane, cyclohexane or cycloheptane, or other not interfering media because they do not interfere with the polymerization process of the olefins.

Further, it is also possible to us a mixture of any of these hydrocarbons.

The first polymerization is preferably carried out in a liquid phase. The advantage of carrying out the first polymerization in a liquid phase is that the reactor is relatively simple to control, and that a fairly good heat transfer takes place.

Preferably, the first polymerization is carried out at a temperature of about 75^0 to 110^0C and a pressure of 40-90 bar. Between these temperature and pressure values the best yield is obtained when a liquid phase is used.

In a preferred embodiment of the present invention the first polymerization of olefins is carried out under supercritical conditions, wherein the polymerization temperature and pressure

are above the corresponding critical points of the mixture
formed by the olefins, catalyst, hydrogen, optional comonomer
and fluidum of inert low boiling hydrocarbon medium and the
polymerization temperature is below the melting point of the
formed polymerized olefins. The temperature and pressure in the
first reactor are preferably about $85°-110°C$ and $60-90$ bar,
respectively.

By using supercritical conditions it is possible to use
higher hydrogen concentrations as would be possible in
subcritical conditions. Further, in supercritical conditions
there is in the reactor practically one single reaction fluid,
where no separate liquid and gas phase can be detected. This
gives very a very good heat transfer, a free mixing of gas
components in the fluid and a low viscosity of the reaction
mixture.

The reactor mixture is removed from the first reactor
either continuously or intermittently. Preferably, hydrogen,
unreacted reactants and inert low boiling hydrocarbon medium are
removed from the reaction mixture. The removal of hydrogen, low
boiling hydrocarbon is preferably carried out by flashing means.

After the reaction mixture has been removed from the first
reactor and hydrogen, unreacted reactants and inert low boiling
hydrocarbon medium are also removed, the polymerized olefins are
fed into the second reactor.

In this second reactor the polymerized olefins are fed into
a fluidized bed and in a moving bed. In the fluidized bed and
the moving bed the residence times may be controlled and
adjusted relative to the total process residence time
independently. During the total residence time of the process
the polymeric particles are recycled during a number of cycles.

The number of cycles in the fluidized bed and in the moving bed
may be selected as desired with the present process according to
the invention. Thus, it is possible to build up polymeric
particles with the desired number and thickness of polymeric
layers formed consecutively in the fluidized bed and moving bed.

According to a preferred embodiment is the residence time
in the moving bed independently controlled, for instance by
controlling the amount of polymeric particles entering the
moving bed but more preferably by controlling the outflow of
polymeric particles out of the moving bed. The control of
outflow rate may have the form of adjusting the opening in the
outlet of the moving bed.

In the moving bed are the catalytic polymerization
conditions different compared to those in the fluidized bed.
First, because the catalytic particles have a higher density in
the moving bed. Furthermore, in order to apply different
polymerization conditions use is made of a separation fluidum in
order to create different catalytic polymerization conditions.
For example, a polymerization carried out at a lower
concentration of a chaingrowth terminating agent such as
hydrogen. Applying a separating fluidum to the moving bed
results in a separation in reaction conditions between the
fluidized bed and the moving bed thereby in the residing
polymerization conditions in the fluidized bed and the moving
bed. Preferably the separation fluidum is added to the top of
the moving bed and forms a cushion on the moving bed through
which cushion of separating fluidum the particulate polymeric
material settles on the forming moving bed. The separation
fluidum may be a gas or a liquid. The separation fluidum may be

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inert to the catalytic polymerization such as nitrogen and C_1-C_{12}-alkane.

The separation fluidum may be reactive such as monomer, comonomer such as C_2-C_{12}-alkylene or mixtures thereof. Mixtures of inert and catalytic polymerization reactive separation fluidum may be used as desired.

Preferably, use is made of a separation fluidum which is a liquid which evaporates under the conditions residing during the catalytic polymerization in the moving bed. Accordingly, during evaporation a gas cushion of separating fluidum is formed and at the same time a cooling of the exothermic polymerization reaction occurs with at the same time a much higher reactant concentration when using reactive separation fluidum.

The addition of separation fluidum but also the reactant to both fluidized bed and moving bed may be such that in the fluidized bed and/or in the moving bed a condensed mode polymerization occurs which is beneficial to productivity and selectivity.

It is further preferred when the separation fluidum comprises a polymerization monomer or comonomer or mixture thereof.

In order to be able to add a rubber component to the polyolefins formed in the second polymerization, it is preferred to carry out a third polymerization in a third reactor. This is preferably carried out in a gas phase reactor. It is even more preferred if in the third reactor the polymerized olefins are further polymerized in a fluidized bed and in a moving bed such that the residence time in the fluidized bed and the residence time in the moving bed are independently controlled.

Before the first polymerization in the first reactor takes
place, a pre-polymerisation is preferably carried out. In such a
pre-polymerisation step the activity of the catalyst is
controlled.

Another aspect of the present invention relates to a
reactor system for the catalytic polymerization of olefins
comprising a first polymerization reactor for carrying out the
first polymerization, which first reactor comprises inlets for
olefins, catalyst, hydrogen, optional comonomer, and inert low
boiling hydrocarbon medium, the first reactor further comprises
a product outlet for a reaction mixture comprising polymerized
olefins; and wherein the product outlet of the first reactor is
connected to an inlet of a second reactor for carrying out the
second polymerization, which second reactor comprises a reactant
inlet, a fluidized bed unit, a moving bed unit and a product
outlet, wherein the fluidized bed unit comprises means for
maintaining a fluidized bed in the fluidized bed unit and
wherein the moving bed unit is provided with an inlet directly
connected to the fluidized bed unit such that the residence time
in the fluidized bed unit and the residence time in the moving
bed unit are independently controlled.

A first advantage of this reactor system is that it is
possible to provide a variety of polyolefins, such as monomodal
and bimodal products with a desired polydispersity and/or
biomodaltiy. A second advantage is that it is possible to
control the built up of the polyolefin particles, such that a
desired number and thickness of the polymeric layers is
obtained.

The first reactor preferably comprises a loop reactor. A
loop reactor has several advantages over other reactors. The

most important advantages of a loop reactor is that it has a
better heat transfer from the reaction mixture and therefore a
more even temperature profile, a higher slurry density and
consequently a better mixing. An other advantage of a loop
reactor is that it permits shorter residence times and as a
consequence the catalyst has not time to inactivate
considerably. This means that when the catalyst is transferred
from the loop reactor to the second reactor it is still very
active.

The loop reactor is preferably adapted such that it can
work under supercritical conditions. By using supercritical
conditions it is possible to use higher hydrogen concentrations
as would be possible in subcritical conditions. Further, in
supercritical conditions there is in the reactor practically one
single reaction fluid, where no separate liquid and gas phase
can be detected. This gives very a very good heat transfer, a
free mixing of gas components in the fluid and a low viscosity
of the reaction mixture.

In order to remove hydrogen, unreacted reactants and inert
low boiling hydrocarbons from the polymerized olefins, is the
product outlet of the first reactor preferably connected to
removal means. These removal means comprise an outlet for the
polymerized olefins that is connected to the inlet of the second
reactor. The removal means preferably comprise flashing means.

The moving bed unit is connected to the fluidized bed such
that a desired part or all fluidized bed reactant is passed
through the moving bed unit. The residence time in the moving
bed unit is controlled by controlling the outflow rate at the
outlet of the moving bed unit. When the moving bed unit is
filled with particulate polymeric material then no further

particulate material form the fluidized bed unit may be added to the moving bed unit and is to be recycled in the fluidized bed unit. The extent of fluidization and filling of the fluidized bed unit determines the residence time of the particulate polymeric material in the fluidized bed unit prior to changing over into the moving bed unit. In the moving bed unit the residence time is dependent on the outflow rate and the particulate polymeric materials follows one cycle within the moving bed unit after entering it.

When in the fluidized bed unit and in the moving bed unit different polymeric reaction conditions reside, then the polymeric particle formed will comprise layers of different polymeric material due to the different polymeric material formed on the particle.

In order to independently control the residence time in the fluidized bed unit and in particular in the moving bed unit, the inlet of the moving bed unit is arranged in the fluidized bed unit so that part of the material from the fluidized bed unit may be transferred into the moving bed unit whereas the other remaining part is further fluidized and mixed in the fluidized bed unit. Generally, the material leaving the moving bed unit is transferred into the fluidized bed unit and ultimately removed from the fluidized bed unit. However, product may also be removed from the moving bed unit.

Various orientations of the fluidized bed unit and of the moving bed unit are contemplated while still the residence time in the fluidized bed unit and in the moving bed unit may be controlled independently. In one embodiment the moving bed unit is arranged within the fluidized bed unit. In an other embodiment the moving bed reactor is arranged around the

fluidized bed reactor and has a substantially annular configuration. In another embodiment, the moving bed reactor is arranged adjacent or even outside the fluidized bed unit while its inlet and optionally its outlet are arranged in the fluidized bed unit. Preferably (as discussed above), the moving bed reactor is provided at least at its inlet with means for supplying the separation fluidum. This separation fluidum is preferably a gas or a liquid and selected from the group comprising an inert gas or liquid, such as nitrogen and C_1-C_{12}-alkene, or C_1-C_{12}-alkylene.

The moving bed unit may be provided with a diverging section such that as from its inlet the unit volume increases thereby allowing a better separation of particulate material entering the moving bed form material which is recycled in the fluidized bed. In another embodiment, the moving bed unit may be provided with a converging section as from its inlet providing a more stable fluidized bed around the moving bed unit

In order to control residence time in the moving bed unit, its outlet is preferably provided with means for controlling the outflow rate of particles from the moving bed. Those outflow rate controlling means may have the form of a valve or any form of construction element suitable for controlling the outflow rate.

The reactor system according to the invention preferably comprises a pre-polymerisation unit connected to the catalyst inlet of the first polymerization reactor.

In a preferred embodiment the reaction system further comprises a third reactor for carrying out a third polymerization, which third reactor is connected to the second reactor. The third reactor is preferably a gas phase reactor. It

is even more preferred if the third reactor comprises a reactant
inlet, a fluidized bed unit, a moving bed unit and a product
outlet, wherein the fluidized bed unit comprises means for
maintaining a fluidized bed in the fluidized bed unit and
wherein the moving bed unit is provided with an inlet directly
connected to the fluidized bed unit such that the residence time
in the fluidized bed unit and the residence time in the moving
bed unit are independently controlled. The preferred embodiments
of the third reactor may be the same as the preferred
embodiments of the second reactor.

The reactor system according to the invention preferably
also comprises a pre-polymerisation reactor. In such a pre-
polymerisation step the activity of the catalyst is controlled.

When producing polypropylene the first polymerization
reactor may comprise propylene as the medium and further if
desired comonomer(s), hydrogen and conventional inerts.

The present invention further relates to the use of a
reactor system according to the invention for the polymerization
of polyolefins.

An other aspect of the present invention relates to
polyolefins obtainable by the process according to the
invention.

A final aspect of the present invention relates to the use
of the polyolefins obtained with the process according to the
invention in pipes, blow molded articles, coating of metal
pipes, jacking of cables, extrusion coating, films and
insulation layers of cables.

Mentioned and other features and advantages of the process
and the reactor system according to the invention are further
explained by way of several embodiments given for illustrative

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purposes and without the intention to limit the invention
thereto, while making references to the figures. In the figures
are:

figure 1 a schematic diagram of the reactor system
according to the invention;

figures 2-6 a schematic diagram of the second reactor used
in the reactor system according to the invention;

figures 7-16 molecular weight distributions obtained with a
process exemplified in the examples 1,2,3 and 4, respectively.

Figure 1 shows a reactor system according to the invention
for the catalytic polymerization of olefins. Olefins used in the
process according to the invention comprise C_2-C_{12}-olefins,
preferably C_2-C_8-olefins such as ethylene, propylene, butene-1,
pentene-1, hexene-1, 4-methyl, pentene-1 and octene. These
olefins may also be used for the production of homopolymers.
Copolymers may be produced by using for instance ethylene and/or
propylene in combination with other C_2-C_8-alpha-olefins. Polyenes
may be used for copolymerization, such as dienes, for instance
1,4-butadiene, 1,6-hexadiene, dicyclopentadiene, ethylidene
norbornene and vinyl norbornene.

For this catalytic polymerization Ziegler-Natta-catalysts
are used which are conventional catalysts available in the are
and selected and desired.

The reactor system 60 comprises a pre-polymerisation
reactor 61 for the pre-polymerisation of the catalyst. The
outlet 62 of the pre-polymerization reactor 61 is connected to
the inlet 64 of the first polymerization reactor 63. The
polymerization comprises a reaction chamber 65 which is adapted
to work under supercritical conditions. The outlet 66 of the
polymerization reactor 63 is connected to flashing means 67 for

separating the polymerized olefins from the inert low boiling
hydrocarbon medium, unreacted reactants and hydrogen. The
flashing means are connected to a second polymerization reactor
68. This second polymerization reactor 68 comprises a fluidized
bed unit 2 and a moving bed unit 3. The outlet of the second
polymerization reactor 68 is optionally connected to the inlet
of a third polymerization reactor 69. Optionally comonomer may
be added to the reactor 61 and/or reactor 63.

Figure 2 shows the second polymerizing reactor 68 according
to the invention, this reactor is described in the European
patent application EP03076791 is incorporated by cross-
reference. The second polymerizing reactor 68 comprises a
fluidized bed unit 2 within which is centrally arranged a moving
bed unit 3.

Monomer is entering the fluidized bed unit via line 4 and
optionally via line 5 inert gas, comonomer and/or hydrogen.

Via a lower space 6 and a gas distribution plate 7 the
reaction gas enters the fluidized bed unit 2. Via line 8 polymer
of the previous polymerization is added from a unit 9 via line
10 and other catalyst components via line 11, and optionally
catalyst compounds may be added.

Sidewise around the moving bed unit 3 is generated and
maintained a fluidized bed of catalytic particles on which
particles due to the polymerization reaction polymer is formed.
The composition of the polymer formed is dependent in the
residing concentration of the monomer, comonomer, inert and
hydrogen. The maintained fluidized bed is visualized by the
arrows 12. At the top of the fluidized bed reactor particles are
separated from gas in a disengaging section 13. Gas leaves the
fluidized bed unit via the recirculation line 14, is optionally

cooled in a heat exchanger 15. compressed in a compressor 16 and
optionally after further cooling in heat exchanger 17 re-added
to the fluidized bed unit 2 via its inlet 18.

Part of the particulate material in the fluidized bed unit
2 visualized by arrows 19 enter the moving bed unit and forms a
bed of settled polymeric particles which move slowly downwardly
in a more or less plug stream towards the outlet 21 which is
partly closed off by a valve 22 control via a control line 23.
On the downwardly moving bed of polymeric particles in the
moving bed 3 is formed a cushion of separation fluidum which is
added via the line 24 and the nozzles 25. This separation
fluidum is in this case a liquid which evaporates under the
conditions residing at the top of the moving bed. The top part
of the moving bed, above the nozzles 25 is preferably fluidized
by upstreaming evaporated separation fluidum.

After a sufficient total residence time in both the
fluidized bed unit 2 and the moving bed unit 3, polymer is
removed via the outlet 25 provided with a valve 26. In a
separator 27 polymeric material is separated and removed via
outlet 28 whereas gaseous material is recycled via line 29 and
line 14.

Figure 3 shows another embodiment of the second
polymerization reactor 68 according to the invention comprising
the fluidized bed unit 2 and moving bed unit 31 centrally
arranged within the fluid bed unit 2. The moving bed unit 31 has
a first diverging upper section 32 and a second converging
section 33.

Due to the diverging section 32 and converging section 33
is formed a complementary structure in the fluidized bed unit
whereby is formed a constrainment 34 dividing the fluidized bed

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in a lower section 35 and upper section 36 which is beneficial
to the stability of the fluidized bed formed.

In the reactor 37 shown in figure 4 the moving bed reactor
38 only has the form of a cup shape whereby more material form
the fluidized bed unit 2 is transferred into the moving bed unit
38.

In the reactor system 39 as shown in figure 5, the moving
bed reactor is formed between an outer wall 40 and a cylindrical
inner wall 41, so that the moving bed unit 42 has the form of an
annular or hollow cylinder with the fluidized bed unit 43
centrally arranged. The outlet 44 of the moving bed unit 42 is a
constrainment in the form of a narrow annular slot trough which
in a defined outflow rate particulate material following the
arrow 45 flows back into the fluidized bed unit 43.

Finally, the reactor system 46 as shown in figure 5
comprise a separate cylindrical moving bed unit 47 arranged
outside the fluidized bed unit 48 and is connected to it via an
inlet 49 and an outlet 50. The outlet 50 is provided with a
control valve 51 for controlling the outflow particulate
material form the moving bed reactor into the moving fuidized
bed unit.

All the reactor types shown in figures 2-6 provide
substantially the same polymer quality if they are operated
under similar conditions. Under nearly isothermal conditions,
beside typical parameters like temperature, pressure, gas and
solid phase composition, "similar conditions" can be defined by
the following major variables:

a)   average residence time of solid and gas phase in
        the whole system

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b)   ratio (average fluid bed unit residence time) : (average moving bed residence time)

c)   average number of powder cycles through fluidized bed and moving bed per average overall residence time

d)   amount of injected liquid

e)   ratio (amount of solids in fluid bed unit) : (amount of solids in moving bed unit)

The fluidization behaviour is influenced by the fluid bed unit and moving bed unit design but is controllable in a wide range of geometric variables.

The results of the following examples did not substantially depend on the specific design when the reactor was operated under "similar conditions". Wall sheeting was never observed. Temperature gradients were always small under all conditions applied.

Examples

A pilot plant was used with about 500 L fluidized bed and 250 L moving bed volume respectively. The mass flow from the first polymerization (homopolymerization loop reactor, see reactor 63 in figure 1) was kept constant in all experiments at about 100 kgh^{-1}, which was controlled by feeding of a controlled amount of a MgCl2 supported Ziegler catalysts, about 30 grams/hour. The molecular weight distribution of the homopolymer is shown in figure 7.

The weight average molecular weight of the feed was 41 kg mol^{-1} with a polydispersity of PD=7.5. The main reactor was operated at constant pressure, P = 25 bar, and constant temperature, T = 85°C. The polymer mass flow of the moving bed (= recycle flow) was adjusted at R = 500 kg h^{-1} and was kept constant within the operation limits.

The following variables were varied during the experiments:

1. molar ratio $X_{C4,1}$ in moles butene per mol ethylene in the fluidized bed unit
2. molar ratio $X_{C4,2}$ in moles butene per mol ethylene in the moving bed unit
3. molar ratio $X_{H2,1}$ in moles hydrogen per mol ethylene in the fluidized bed unit
4. molar ratio $X_{H2,2}$ in moles hydrogen per mol ethylene in the moving bed unit
5. internal recycle ratio

Example 1
The following conditions were applied:

$X_{C4,1}$= $X_{C4,2}$=1

$X_{H2,1}$= $X_{H2,2}$=0.045

The reaction conditions in both reactor unit do not differ significantly. It can be assumed that both reactor units produced nearly the same polymer quality. 121 kg copolymer per hour was produced in this experiment with a butene content of 2.5 mol% and an average molecular weight of 246 kg mol^{-1}. With 500 kg per hour polymer flow in the recycle, the recycle ratio is about 4. The polydispersity of the molecular weight

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distribution, MWD shown in fig. 8, is 6.55. The polymer was
analyzed by means of a TREF method to estimate the number of
butene units per 1000 carbon atoms as function of the molecular
weight. This value was multiplied with the amount of polymer
produced, which exhibits the given butene units, to get the
"chemical distribution" demonstrated in figure 9. It was found
that the low molecular weight polymer contains more comonomer
than the high molecular weight part under these conditions,
which can be seen as a specific catalyst property.

Example 2
The following conditions were applied:
$X_{C4,1}= X_{C4,2}=0.5$
$X_{H2,1}= 0.3;$ $X_{H2,2}=0.005.$
This example is to demonstrate the broadening of the molecular
weight distribution keeping the chemical gas composition in both
reactor units nearly the same. Usually, with this type of
catalyst, which produces a broad molecular weight distribution
even under constant reaction conditions, it is difficult to
produce a bimodal MWD. However, with the reactor described here
it was possible to broaden the MWD remarkably by just keeping H2
away from the moving bed. The broad high molecular weight
shoulder, see fig. 10, tends to bimodality. Interestingly,
nearly the same polymer production as in example 1 resulted, 120
kg per hour. The average chemical composition was 1.43 mol%; the
weight average molecular weight is 202 kg per mol; the
polydispersity was large, PD= 14.8 was observed.

The peak of the chemical distribution, CD, is lower, corresponding to the lower average comonomer content, but the whole CD is broadened preferrably by the influence of the MWD.

Example 3

The following conditions were applied:

$X_{C4,1}= 0.1$; $X_{C4,2}=1$

$X_{H2,1}= 0.3$; $X_{H2,2}=0.005$.

In this experiment, was obtained a similar polydispersity of the MWD as in example 2, PD=14.6, and also the MWD itself looks similar with the enlarged high molecular weight shoulder, but only 105 kg per hour was produced with an average chemical composition of (similar to example 2) 1.58 mol%. Interestingly, by comparing fig. 10 and fig. 13, this time the high molecular weight shoulder is enriched with the comonomer which increases the desired product quality strongly.

These examples have shown that manipulation of both MWD and CD is possible in the second polymerization by means of common Ziegler catalysts, just by controlling the gas composition in both reactors independently.

Example 4 demonstrates the influence of the independently controlled recycle ratio based on results of example 3.

Example 4

The following conditions were applied:

$X_{C4,1}= 0.1$; $X_{C4,2}=1$

$X_{H2,1}= 0.3$; $X_{H2,2}=0.005$

R = 1350 kg h^{-1} (internal recycle flow).

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The increased recycle flow in the moving bed unit led to again improved comonomer incorporation, see fig. 15, at slightly decreased polydispersity, which is PD=13.2 now. At the same time, the average comonomer composition increased to 1.76 mol% as well as the weight average molecular weight reached M_w=303 kg mol^{-1}. Positive also the polymerization rate change: 126 kg per hour was observed.

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Legend to the Figures 7 to 15

Figure 7: Molecular weight distribution of the homopolymer
polymer from reactor 1.

Figure 8: Molecular weight distribution, final product, example
1.

Figure 9: Chemical distribution, final product, example 1.

Figure 10: Molecular weight distribution, final product, example
2

Figure 11: Chemical distribution, final product, example 2.

Figure 12: Molecular weight distribution, final product, example
3.

Figure 13: Chemical distribution, final product, example 3.

Figure 14: Molecular weight distribution, final product, example
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Figure 15: Chemical distribution, final product, example 4.

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